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A Search for Interstellar CH_3D: Limits to the Methane Abundance in Orion-KL

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Abstract

A search has been performed for interstellar CH_3D via its $J_K = 1_0 - 0_0$ transition at 230 GHz and its $J_K = 2_0 - 1_0$ and $J_K = 2_1 - 1_1$ lines at 465 GHz using the NRAO 12 m and CSO 10 m telescopes towards Orion-KL. This search was done in conjunction with laboratory measurements of all three transitions of CH_3D using mm/sub-mm direct absorption spectroscopy. The molecule was not detected down to a 3σ level of $T_A^* < 0.05$ K towards Orion, which suggests an upper limit to the CH_3D column density of $N < 6 \times 10^{18}$ cm^{-2} in the hot core region and a fractional abundance (with respect to H_2) of $< 6 \times 10^{-6}$. These measurements suggest that the methane abundance in the Orion hot core is $f < 6 \times 10^{-4}$, assuming $D/H \sim 0.01$. Such findings are in agreement with recent hot core chemical models, which suggest $CH_4/H_2 \sim 10^{-4}$.

Subject Headings: ISM: abundances, ISM: clouds, ISM: molecules - molecular processes

1. Introduction

Chemical models predict that CH_4, the simplest stable organic molecule, to be one of the most abundant polyatomic species in interstellar dense clouds, since it only involves one carbon atom bonded to four hydrogen atoms (e.g. Mitchell 1977, Brown & Rice 1986). In addition, methane is thought to have an enhanced abundance in hot, dense gas, such as the Orion hot core region due to evaporation of this molecule from grain mantles (e.g. Brown, Charnley, & Millar 1988). However, CH_4 is completely symmetric, having the geometry of a perfect tetrahedron, and hence does not possess a permanent electric dipole moment. Thus, the molecule does not have a normal pure rotational spectrum and consequently cannot be readily observed in cooler, dense gas typical of molecular clouds.

Methane, on the other hand, does possess a rovibrational spectrum in the infrared which has been detected towards IRC+10216 (Hall and Ridgway 1978) and possibly in a few star-forming regions (Lacy et al. 1991). However, the kinetic temperatures found in molecular clouds, even toward star-forming regions, are typically only $T \sim 10$-100 K. Therefore, vibrational excitation is not commonly found in interstellar molecules, especially for regions of extended gas, the dominant constituent of molecular clouds. Hence, it is difficult to test models of interstellar chemistry using CH_4 infrared observations.

An alternative method to obtain interstellar methane abundances is possibly by observing its singly-deuterated form, CH_3D, which has a pure rotational spectrum. Because CH_3D is only slightly asymmetric, however, its dipole moment is small: $\mu \sim 0.006$ D (Wofsy, Muenter, & Klemperer 1970). Moreover, given the normal cosmic D/H ratio of 10^{-5}, detection of CH_3D should prove difficult. A search was carried out for CH_3D via its $J_K = 1_0 - 0_0$ transition at 232 GHz by Pickett, Cohen, and Phillips (1980), but given the receiver technology at the time, not very sensitive limits ($T_A^* \lesssim 0.2$ K) were obtained in Orion-KL. However, recent observations of interstellar molecules have clearly demonstrated that significant deuterium fractionation may occur in dense clouds and that the cosmic D/H ratio has little to do with abundances of deuterated species in comparison with their parent molecules. For example, considerable enrichment is found in several deuterated molecules toward Orion-KL, with $HDCO/H_2CO \sim 0.01$-0.03, $CH_3OD/CH_3OH \sim 0.01$-0.06, and $NH_2D/NH_3 \sim 0.003$-0.05 (Loren and Wootten 1985; Mauersberger et al. 1988; Walmsley et al. 1987).

Here we present results of a renewed search for interstellar CH_3D towards Orion-KL. We have conducted measurements of the $J_K = 1_0 - 0_0$ transition at 232 GHz and the $J_K = 2_1 - 1_1$ and $2_0 - 1_0$ lines at 465 GHz of this isotopomer. We also have carried out laboratory measurements of all three CH_3D transitions. Although spectral lines were detected at the $J_K = 1 \rightarrow 0$ and $J_K = 2_0 - 1_0$ frequencies in Orion, no features were present at the $J_K = 2_1 - 1_1$ line, hence making our search negative. In this paper both the astronomical and laboratory data are summarized. We also present upper limits for the methane abundance and discuss its implications for interstellar chemistry.

2. Observations

The fundamental transition $J_K = 1_0 - 0_0$ of CH_3D at 232,463.3 MHz (see Pickett, Cohen, and Phillips 1980) was originally searched for using the NRAO[1] 12 m telescope at Kitt Peak, AZ in April 1991. At this frequency, the beam size was 28" and the beam efficiency was $\eta_B = 0.4$. The temperature scale was determined by the chopper wheel method, corrected for forward spillover losses,

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[1] NRAO is operated by the Associated Universities, Inc., under contract with the National Science Foundation.

and is given in terms of T_R^*. Conversion to radiation temperature is then $T_R = T_R^*/\eta_B$. The receiver used was a dual channel SIS mixer. The backends used were two 256 channel filter banks with 1 MHz resolution, one for each receiver channel. Data was taken in a double sideband (DSB) mode.

Additional observations were done of the $J_K = 1_0 - 0_0$ line using the Caltech Submm Observatory (CSO) in 1993 September. At 232 GHz, $\theta_b \sim 40''$ and $\eta_B = 0.8$. The receiver used was a single channel SIS mixer, again operated in DSB mode. In addition, measurements of the two K components of CH_3D near 465 GHz were conducted 1992 March and 1995 February, again using the CSO. The two K-components were observed simultaneously. The beam size at this frequency is $\theta_b \sim 15''$ and $\eta_B = 0.5$. Again, the receiver used was a single channel SIS mixer. The temperature scale for all CSO observations is in terms of T_A^*, such that $T_R = T_A^*/\eta_B$. The backend used was a 1024 channel AOS spectrometer with 500 kHz resolution. All observations were carried out towards the position $\alpha = 5^h32^m46^s8$; $\delta = -5°24'23''.0$ (1950.0) in Orion.

3. Experimental

Spectra of CH_3D were obtained in the laboratory using a millimeter/sub-milliliter wavelength direct absorption spectrometer, which is described in detail elsewhere (Ziurys et al. 1994). Briefly, the instrument consists of a tunable source of mm/sub-mm radiation, a gas absorption cell, and a detector. The source for the spectrometer are phase-locked Gunn oscillators (65-140 GHz), used in conjunction with frequency multipliers to cover the range 115-520 GHz. The radiation is quasi-optically propagated through the absorption cell, which is a double-pass system. The detector is a helium-cooled InSb bolometer.

For these experiments, CH_3D gas, purchased from a commercial source (Icon), was sealed in a quartz glass tube about 6 cm in diameter and 400 m in length with quartz flats sealing both tube ends. CH_3D was added to the cell at the desired pressure (~50 mtorr) and the cell then sealed. The tube was then inserted into the spectrometer and data taken, allowing for long signal averaging without having to free-flow CH_3D gas.

4. Results

In the initial search for CH_3D using the NRAO 12 m telescope, a weak feature was found at the frequency of the $J_K = 1_0 - 0_0$ line at 232 GHz towards Orion-KL. This line was subsequently confirmed using the CSO, and this spectrum is shown in the top panel of Fig. 1. As can be calculated from Table 1, this feature has a linewidth of ~4 km/s and an LSR velocity near 5 km/s, if it arises from the $J_K = 1_0 - 0_0$ line of CH_3D. The frequency of this transition of CH_3D had been previously measured in the laboratory by Pickett, Cohen, and Phillips (1980) to be 323,644.327(18) MHz, and hence there was no question as to its exact value. Such line parameters suggested that the observed feature, if due to CH_3D, had an origin in the Orion hot core because of its slightly lower velocity (5 vs. 9 km/s). Because large enhancements for deuterated species are commonly found in the hot core (e.g. Plambeck and Wright 1987), this result was not unexpected. A feature was also detected at near the CH_3D frequency in SgrB2 and W51.

The results of the search for the $J_K = 2_1 - 1_1$ and $2_0 - 1_0$ transitions near 465 GHz were not as clear. In the March 1992 measurements, the data showed two spectral features which possibly could correspond to the $J_K = 2_1 - 1_1$ and $J_K = 2_0 - 1_0$ components of CH_3D, provided they had velocities near 5-6 km/s. However the candidate $J_K = 2_1 - 1_1$ feature appeared as a shoulder on a stronger line, which is unidentified, and the signal-to-noise was not particularly good. Moreover, the frequencies of these two K components of the $J = 2 \rightarrow 1$ transition of CH_3D had never been directly measured before in the laboratory, and could only be predicted from the ro-vibrational data of Tarrago et al. (1976). The absolute accuracy of such calculations was not clear.

Because of these uncertainties, we decided to directly measure the $J = 2 \rightarrow 1$ CH_3D transitions in the laboratory, and remeasure the $J = 1 \rightarrow 0$ line frequency as well. The details of these measurements have been described in the experimental section of this paper. The resulting rest frequencies are listed in Table 2, and have an accuracy of better than ±75 kHz. A sample spectrum of the $J = 2 \rightarrow 1$ transitions is shown in Fig. 2.

With these new laboratory frequencies, we repeated our search for the $J = 2 \rightarrow 1$ lines of CH_3D in the February 1995. For these observations, a spectrum with much better sensitivity was obtained towards Orion because of an improved receiver. These data are shown in the bottom panel of Fig. 1. As this figure illustrates, a line with $T_A^* \sim 0.5$ K is present at the frequency of the $J_K = 2_0 - 1_0$ transition, for a velocity of $V_{LSR} \simeq 5$ km/s, the same LSR velocity as the $J = 1 \rightarrow 0$ candidate line. However, there is no obvious feature present at the frequency of the $J_K = 2_1 - 1_1$ transition, which is 15.15 MHz lower in frequency (or to the left) of the $J_K = 2_0 - 1_0$ component. There are lines near 465,238 MHz and 465,232 MHz, either which could correspond to the $J_K = 2_1 - 1_1$ line, but neither have a matching $J_K = 2_0 - 1_0$ component ~15 MHz to higher frequency.

The obvious failure to detect two distinct K-components of CH_3D is clear evidence for its absence in Orion. The components are separated in energy by only 8 K, so both should be detectable in the high excitation gas of the Orion hot core. An upper limit to the CH_3D antenna temperature can be derived from the $J_K = 2_1 - 1_1$ component, which is $T_A^* \sim 0.05$ K.

The frequencies, intensities, and linewidths of the spectral lines detected in Figure 1 are listed in Table 1. In addition, revised rotational constants for CH_3D

are given in Table 2. As the table shows, they are in good agreement with past estimates from Pickett et al. (1980) and Tarrago et al. (1976).

5. Discussion

5.1 *Column Density and Fractional Abundance for CH_3D*

The upper limit to the CH_3D column density was calculated using the following formula, which assumes low optical depth, for $(J+1)_K \rightarrow J_K$:

$$N_{tot} \leq \frac{3k10^5 T_R \Delta v_{1/2} J \cdot e^{h\nu/kTex} Q_{rot}}{8\pi^3 \nu \mu_0^2 (J^2 - K^2) e^{-\Delta E/kTrot} S_{I,K}} \qquad (1).$$

In this equation, which does not assume the Rayleigh-Jeans approximation, ν is the frequency of the transition, μ_o the dipole moment, ΔE the energy of the J^{th} level above ground state, and T_{rot} and T_{ex} the rotational and excitation temperatures, respectively. T_R is the upper limit to the measured line temperature, and $\Delta v_{1/2}$ the assumed linewidth. The term Q_{rot} is the rotational partition function, which for a symmetric top molecule can be approximated by (e.g. Townes and Schawlow 1975):

$$Q_{rot} \sim \sqrt{B^2 C / \pi T_{rot}} \qquad (2),$$

where B and C are the rotational constants of the molecule. $S_{I,K}$ defines the statistical weight factor due to the presence of three equivalent nuclei with $I = 1/2$, i.e. the three protons (also see Townes and Schawlow 1975):

$$S_{I,K} = \frac{2(4I^2 + 4I)}{4I^2 + 4I + 1} \qquad (3).$$

In this formula, the 2.7 K microwave background was neglected (c.f. Ziurys, Hollis, and Snyder 1994). Also, because the dipole moment of CH_3D is so small, it was assumed that $T_{ex} = T_{rot}$, i.e., the excitation temperature within the 2_1-1_1 transition equals that governing the population in the rotational ladder.

To actually calculate the limit to the column density, it was assumed that CH_3D would arise primarily from the Orion hot core region. Simple, heavily saturated species such as NH_3 appear to high abundances in the hot core, and CH_4 is likely to follow this pattern (e.g. Brown, Charnley and Millar 1988). For this region, the approximate kinetic temperature is ~ 200 K (e.g. Blake et al. 1987), and hence it was assumed that $T_{ex} = T_{rot} = 200$ K. Also, $\Delta v_{1/2}$ was estimated to be 10 km/s, the canonical "hot core" linewidth. The upper limit to the line radiation temperature was $T_R = T_A^* / \eta_B = 0.1$ Km, where $T_A^* < 0.05$ was the upper limit for the $J_K = 2_1 - 1_1$ transition.

Using these assumptions, the upper limit to the CH_3D column density in the Orion hot core was calculated to be $N_{tot} \lesssim 2 \times 10^{18}$ cm^{-2}. This number is based solely on the limit obtained for the $J_K = 2_1 - 1_1$ transition (assuming $V_{LSR} = 5$ km/s), which was not contaminated by other lines as the other two transitions were. This limit assumes the source fills the 15" beam at 465 GHz. Correcting for the hot core size of 10", (Masson et al. 1985), however, increases the upper limit by a factor of 3, or $N_{tot} < 6 \times 10^{18}$ cm^{-2}. If the total hydrogen column density is assumed to be $N(H_2) \sim 10^{24}$ cm^{-2} (e.g. Masson et al. 1985), as is appropriate for the hot core, then the fractional abundance of CH_3D in this region is $f \lesssim 6 \times 10^{-6}$.

5.2 *Implications for the CH_4 Abundance.*

Estimating the methane concentration from CH_3D depends critically on the D/H ratio, which fortunately, can be estimated from other molecules in Orion. Large deuterium enhancements have been found in several clumps towards the

KL/IRc2 region, including the hot core, the so-called "compact ridge", and in the "northern condensation" (eg. Mangum, Plambeck, and Wootten 1991). In the hot core, measurements of NH_2D/NH_3 by Walmsley et al. (1987) yield D/H ~ 0.003, while DCN/HCN suggest D/H ~ 0.005 (Mangum, Plambeck, and Wootten 1991). Observations of CH_3OH and CH_3OD by Mauersberger et al. (1988) indicate higher ratios of D/H ~ 0.01-0.06. However, it is not entirely clear from what region the CH_3OD lines actually arise. If a D/H ratio of 0.01 is assumed, the upper limit to the fractional abundance of methane in the Orion hot core is $f \lesssim 6 \times 10^{-4}$, with a corresponding column density limit of N_{tot} $(CH_4) \lesssim 6 \times 10^{20}$ cm^{-2}.

The only source where methane has conclusively been detected is in the circumstellar shell of IRC+10216 (Hall and Ridgway 1978). These authors observed the 3.3 μm v_3 vibrational band of CH_4, and derived a corresponding column density of $N_{tot} = 3 \times 10^{17}$ cm^{-2}. They also measured a CO column depth of $N_{tot} = 10^{20}$ cm^{-2} in IRC+10216. If $CO/H_2 \sim 10^{-4}$, then the fractional abundance of methane, relative to H_2, is $f \sim 3 \times 10^{-7}$ in this object. This value is several orders of magnitude below our upper limit of $f \lesssim 6 \times 10^{-4}$ in Orion. However, the chemistry in the envelope of IRC+10216, a late-type carbon star, is quite different from that of the Orion hot core, so methane abundance are likely to vary between the two sources.

Lacy et al. (1991) searched for the 7.6 μm v_4 band of CH_4 towards several molecular clouds. There authors appeared to detect the R(0) and R(2) lines of this band towards NGC7538 IRS9, although the spectra were highly contaminated by telluric features. From these two lines, Lacy et al. derived a CH_4 column density of $N_{tot} \sim 2 \times 10^{16}$ cm^{-2} towards NGC7538. Using their quoted CO column depth of 1.5×10^{19} cm^{-2}, and assuming $CO/H_2 \sim 10^{-4}$, the fractional abundance of CH_4 in this molecular cloud is $f \sim 10^{-7}$, relative to H_2. A similar fractional abundance was possibly found for W33A. Again, these CH_4 abundances are several orders of magnitude lower than our upper limit.

Lacy et al. also suggest a possible detection of the R(0) line of methane in Orion/IRc2. However, they do not derive a column density for CH_4 from this measurement, although a comparison of methane abundances may have been useful. Lacy et al. determined an LSR velocity of the possible R(0) feature of V_{LSR} = -1 km/s. To our knowledge there is no known molecular emission in Orion that occurs at that velocity.

5.3 *Constraints for Chemical Models*

The "hot core" cloud model by Brown, Charnley, and Millar (1988) predict high abundances of simple saturated molecules such as NH_3 and CH_4. These calculations basically considered gas phase reactions and grain mantle evaporation in dense gas heated by a young star, with T ~ 150 K. In this model, these authors estimate $f\,(CH_4) \sim 10^{-4}$, relative to H_2. A newer set of calculations by Brown and Millar (1989) basically consider the same model as Brown et al. (1988), but additionally consider some grain-surface reactions. Once again, the estimated abundance of methane for hot core-type gas is $f \sim 10^{-4}$.

The upper limit for methane in the Orion hot core region determined from the CH_3D search suggests $f \lesssim 6 \times 10^{-4}$. This value certainly does not contradict model predictions; however, it is not a stringent enough limit to put the chemical calculations to the test. Given the contaminating spectral features at two of the three easily accessible CH_3D rotational transitions, obtaining more sensitive limits will be difficult.

6. Conclusions

A search has been carried out for the $J_K = 1_0 - 0_0, 2_0 - 1_0$, and $2_1 - 1_1$, mm/sub mm transitions of CH_3D at 232.64, 465.25, and 465.24 GHz. Laboratory measurement of these CH_3D frequencies were also performed to substantiate the astronomical work. Although spectral lines were clearly present at the 1_0 - 0_0 and 2_0 - 1_0 frequencies for CH_3D in Orion, assuming the hot core velocity, there was no obvious feature at the 2_1 - 1_1 frequency. Hence, CH_3D was clearly not detected in this source. The upper limit to the column density and fractional abundance of CH_3D on the Orion hot core, relative to H_2, is $N_{tot} \lesssim 6 \times 10^{18}$ cm^{-2} and $f \lesssim 6 \times 10^{-6}$. These values imply that the methane abundance in the hot core is $f < 6 \times 10^{-4}$, consistent with current chemical models, which predict $CH_4/H_2 \sim 10^{-4}$.

This search for CH_3D also illustrates the need for careful and objective measurements in the detection of new interstellar molecules. Based on two lines alone (the $J_K = 1_0$ - 0_0 and 2_0 - 1_0 transitions), we could have claimed an interesting discovery of interstellar CH_3D. However, failure to observe a consistent third transition, namely the $J_K = 2_1$ - 1_1 line, showed beyond any doubt that such a claim would have been premature and absolutely incorrect.

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Table 1: Lines Observed in the Bandpass

Frequency (MHz)	Identification	T_A^*(K)	V_{LSR} (km/s)	$\Delta V_{1/2}$(km/s)	Comments
229,647.7	VyCN	0.12	5.3	6.5	LSB line; appears twice in spectrum
U232,638	---	~0.1	9.0	~5	blended with VyCN
U232,647	---	0.1	9.0	3.8	near CH_3D; $J_K = 1_0 - 0_0$ transition
U465,232	---	0.5	9.0	~6	blended with U 465,238
U465,238	---	0.6	9.0	~6	blended with U465,232
U465,258	---	0.5	9.0	5.8	near CH_3D; $J_K = 2_0 - 1_0$ transition
U468,120	---	1.8	9.0	4.5	---

Table 2: Observed Frequencies and Revised Constants for CH_3D

Transition (J_K)	Frequency (MHz)
$1_0 - 0_0$	232,644.301 (0.075)
$2_0 - 1_0$	465,250.691 (0.075)
$2_1 - 1_1$	465,235.540 (0.075)

Constant	ν (MHz)[a]	Previous Work (MHz)[a]
B_o	116,325.309(12)	116,323(2)[b], 116,325.308(27)[c]
D_J	1.5725(21)	1.572(7)[b]
D_{JK}	3.79(3)	3.78(3)[b]

a) Errors listed are 3σ.

b) From Tarrago et al. (1976).

c) From Pickett, Cohen, and Phillips (1980).

Figure Captions:

Figure 1: Spectra taken at the frequencies of the CH_3D transitions at 323 and 465 GHz, respectively, using the CSO. The top panel shows the spectrum at 232 GHz, where a spectral line occurs near the CH_3D:$J_K = 1_0 - 0_0$ frequency. A vinyl cyanide line is present in the LSB in these data, which appears twice due to an LO-shift. For one LO-setting, it is blended with a U-line from the USB. In the bottom panel, the data taken at 465 GHz are presented. Although a spectral feature occurs near the $J_K = 2_0 - 1_0$ frequency, there is not a line corresponding to the $J_K = 2_1 - 1_1$ transition. The frequency scale assumes $V_{LSR} = 9$ km/s in both cases.

Figure 2: Laboratory spectrum of the CH_3D:$J_K = 2_0 - 1_0$ and $J_K = 2_1 - 1_1$ transitions near 465 GHz. These measurements confirm the 15 MHz splitting of the two K components of this transition. This spectrum represents one, 3 minute scan.

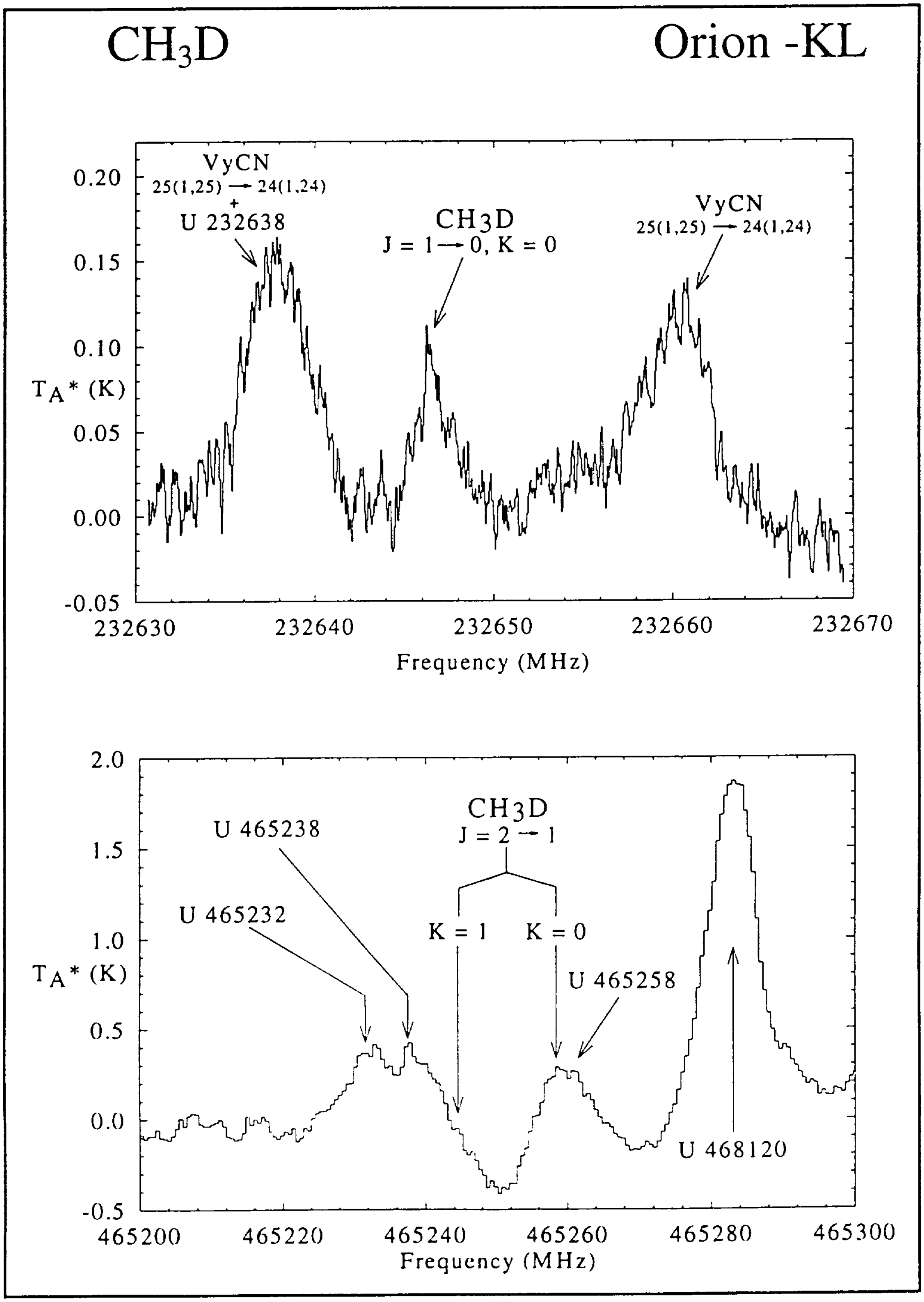
CH3D
Orion -KL
VyCN
25(1,25) → 24(1,24)
+
U 232638
CH3D
J = 1 → 0, K = 0
VyCN
25(1,25) → 24(1,24)
TA* (K)
0.20
0.15
0.10
0.05
0.00
-0.05
232630
232640
232650
232660
232670
Frequency (MHz)
U 465238
U 465232
CH3D
J = 2 → 1
K = 1
K = 0
U 465258
U 468120
TA* (K)
2.0
1.5
1.0
0.5
0.0
-0.5
465200
465220
465240
465260
465280
465300
Frequency (MHz)

Fig. 1

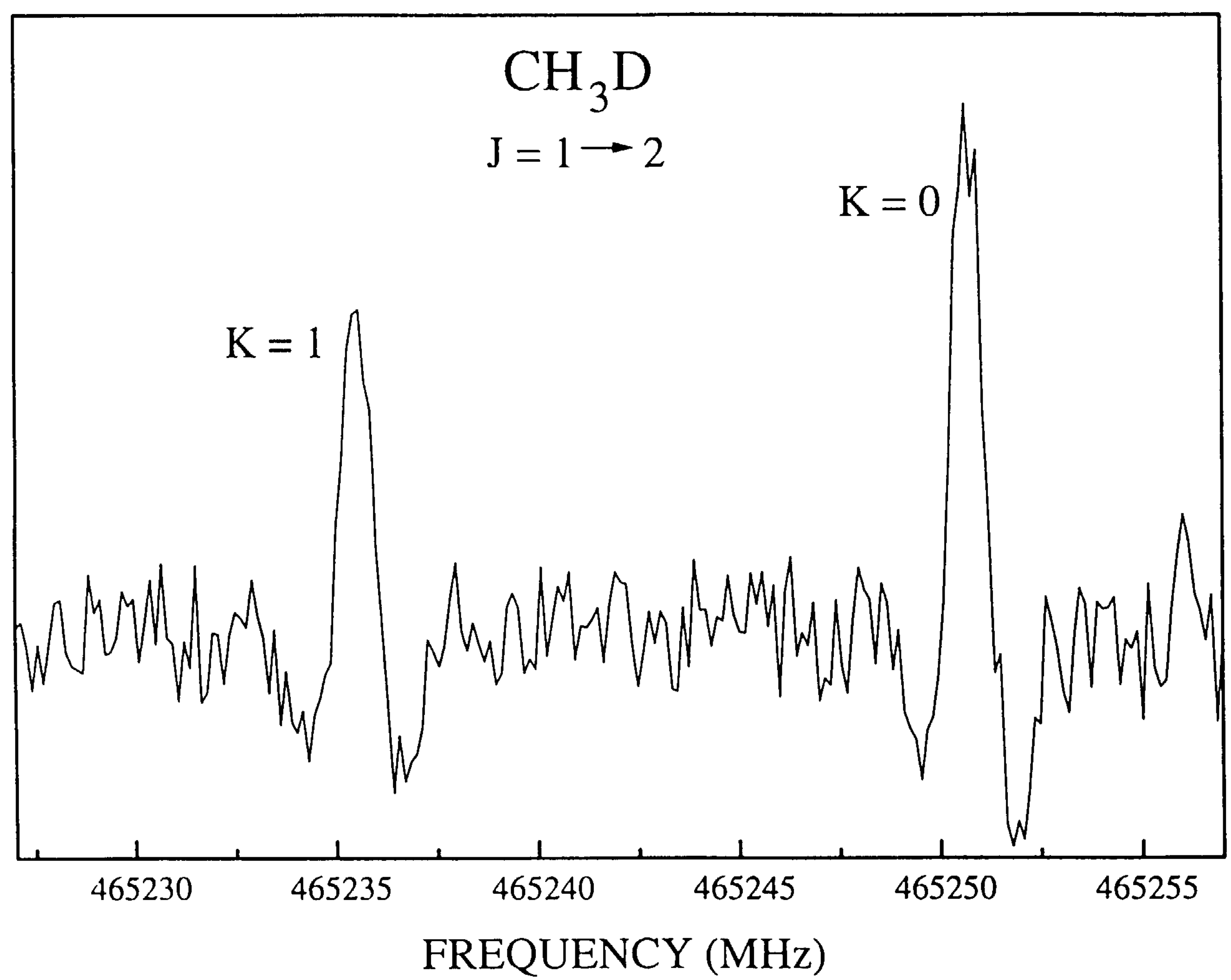
CH3D
J = 1 → 2
K = 0
K = 1
465230
465235
465240
465245
465250
465255
FREQUENCY (MHz)

Fig.2